IN THE UNITED STATES PATENT AND TRADEMARK OFFICE

Applicant: Lieven ANAF et al.

Title: POROUS METAL STACK FOR FUEL CELLS OR
                 ELECTROLYSERS

Appl. No.:   10/501,145

International   12/19/2002
Filing Date:
371(c) Date: 7/13/2004

Examiner: Steven M. Scully

Art Unit: 1795

Confirmation   5358
Number:

<u>DECLARATION UNDER 37 C.F.R. § 1.131</u>

Commissioner for Patents
P.O. Box 1450
Alexandria, VA 22313-1450

Sir:

I, Marc Messely, declare as follows:

1.   I am employed by N.V. Bekaert S.A., the assignee of this application and are submitting the declaration on behalf of the assignee.

2.   The invention of independent claims 1 and 3 of the above-identified application was conceived at least as early as June 12, 2001 and diligence was exercised until an embodiment of the invention was actually reduced to practice at least by September 24, 2001 and that diligence was exercised at least until the European priority application was filed on January 15, 2002.

3.   To demonstrate this earlier date of conception and diligence until the European priority application was filed, the following documents are attached: a notary public registration of an email, a notary public registration of a facsimile transmission, drafts of the European priority application, and handwritten notes on a draft European application.

4.   As shown in the attached notary public registration, Exhibit A, the invention for the above-identified application was conceived at least as early as June 12, 2001. Although the first page of the notary public registration is in English, the second and third pages are in Dutch. The following is an English language translation of the second and third pages, with the second page being a title

page and the third page being an email by co-inventor Lieven Anaf to Wim Van Steenlandt on June 11, 2001 relating to fuel cell developments with Nuvera:

Second page:

Document number 2261
Bearing as feature Fuel Cell - Development
Comprising two pages
Belonging to N.V. Bekaert S.A. at Zwevegem
Signed as "ne varietur" on the first page or envelope and further signed on the other pages by notary public Fr. Opsomer in Kortrijk in the presence of two witnesses on the 12th of June 2001 in order to confirm the existence thereof at that date.
(signatures)

Third page:

Dear Wim,

Last week Nuvera Italy paid a visit. In the discussions it became apparent (our proposal by the way) that we could make for them a fuel cell stack out of a combination of a bipolar plate of stainless steel (316 or 310) sintered to a gas diffuser (e.g. a layer of 30 micron fibers of 316L with 85% porosity) and there above a layer of 8 micron fiber of 316L 70% porosity as electrode layer, sintered to the rough fiber layer. This "fine" layer makes contact with the PEM membrane. One may even conceive the combination fine electrode, rough gas diffuser, bipolar plate, rough gas diffuser, electrode, everything sintered together, which may be mounted between various PEM-membranes during construction of the fuel cell stack. Typical thicknesses are: gas diffuser 1.5 à 2 mm, electrode 0.1 à 0.2 mm, bipolar plate 0.2 mm.

The purpose of the combination is to solve the electrical contact problems (since everything is sintered together): all separate parts are now coated with an electroconductive coating (preferably a noble metal) to minimize the contact resistance. The use of 316L also reduces that material costs and provides corrosion resistance in the given circumstances of 70°C and pH 5, as claimed by Nuvera.

The question is to make at least a registration with a notary public and possibly even a patent, also in the framework of possible questioning to other potential players in the market.

In your search to patents relating to electrodes, I have found one patent which describes about the same (namely layers sintered together), however for batteries, patent from Westinghouse.

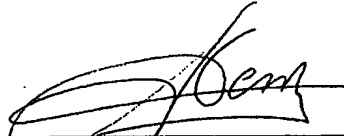What is your opinion, can we sit together?

2

Brgds,

Lieven

5.   The notary public registration demonstrates that the invention of claims 1 and
      3 was conceived of at least by the date of June 12, 2001.

6.   Diligent efforts were undertaken until an embodiment of the invention of
      claims 1 and 3 was reduced to practice and at least until the European patent
      application was filed on January 15, 2002. For instance, on June 22, 2001,
      Nuvera sent specifications for a fiber stack to Bekaert and additional details
      for the fiber stack were developed, including tests of samples. On July 20,
      2001, a sample was tested and it was determined that the porosity of the
      electrode layer and the gas diffusion layer should be as different as possible
      and that four samples should be made.

7.   As shown by Exhibit B, which is another notary public registration of a
      facsimile transmission, four Bekaert fibers stacks were sent to Nuvera. The
      fiber stacks having different layers with different porosities demonstrate that
      an embodiment of the invention of claims 1 and 3 was reduced to practice by
      at least September 24, 2001, the date of the notary public registration.

8.   Furthermore, diligent efforts were continued until at least the European
      priority application was filed. On October 19, 2001, a first draft of the
      European patent application was drafted, a copy of which is attached as
      Exhibit C. On November 8, 2001 a second draft of the European application
      was drafted, a copy of which is attached as Exhibit D. On November 21,
      2001, hand written additions were made to the draft application, a copy of
      which is attached as Exhibit E. On November 22, 2001, a further draft of the
      European application was made, a copy of which is attached as Exhibit F. The
      further draft was mailed to the inventors on December 19, 2001, a copy of
      which is attached as Exhibit G, and the further draft was edited on January 9,
      2002 and January 11, 2002 before it was filed January 15, 2002.

9.   These statements and the attached exhibits demonstrate diligence in the actual
      reduction to practice of an embodiment of the inventions of claims 1 and 3,
      and diligence in the preparation of a draft patent application after conception
      of the invention.

10.   All of the work discussed above and reflected in the content of the attached
      documents was performed in Belgium.

11.   Thus, the inventions of independent claims 1 and 3 was conceived as early as
      June 12, 2001 and diligence was exercised until an embodiment of the
      invention of claims 1 and 3 was actually reduced to practice and until the
      European priority application was filed.

12.   I hereby declare that all statements made herein of my own knowledge are true
      and that all statements made on information and belief are believed to be true;
      and further that these statements were made with the knowledge that willful

false statements and the like so made are punishable by fine or imprisonment, or both, under Section 1001 of Title 18 of the United States Code and that such willful false statements may jeopardize the validity of the application or any patent issued thereon.

Date: _April 21, 2009_

Marc Messely

EXHIBIT B

Form for NOTARIAL REGISTRATION

$\underline{2300}$ 24/09/2001

only to be filled in by the Industrial Property Department :

Number of notarial registration : _2300_

Related file at IPD : _5583_

Date of registration : _24/09/2001_

Title of registration : _Producten gelaverd aan Nuvera_

Name requestor : _L Anal_ ,

Read and understood by (name) : _Wim Van Steenlandt_

       (function) : _Trainee patent Attorney_

       (signature) : _Van Steenlandt_

       (date) : _____ _24/9/01_

following data to be procured by the requestor :

Author of the registered information : _L. Anal_

Place of disclosure of the information :
BFT- Brussel ; Nuvera

Date of communication of the information : _21/9/01_

Receivers of the disclosed information :
Gianpaolo Sibilia
_____

Way of disclosure of the information :
- under an explicit secrecy agreement   (yes) / no
- under an oral request for secrecy   yes / no
- information marked as confidential   (yes) / no
- during a meeting open to the public   yes / no
- distribution of copies with information   yes / no
                  (number of copies : ____)

facsimile transmittal

To:	Giampaolo Sibilia	Fax:	+39 02 21292403
From:	Lieven Anaf	Date:	21/09/01
Re:	samples for fuel cell stack	Pages:	4
CC:	Mr Liotta, Pascal Catteeuw		

☐ Urgent   ☐ For Review   ☐ Please Comment   ☐ Please Reply   ☐ Please Recycl

Dear Mr Sibilia,

Please find herewith the description of the samples sent to you a week ago.

We had a problem with the 4th series of samples, because we used two times a wire mesh, which did not sinter together. We didn't have a wire mesh with the right thickness available.

We would like to receive 6 additional bipolar plates, to redo this test with a thicker wire mesh.

A remark is that we are not able to sinter the wire mesh to the bipolar plate, this means we always will need a fibre layer in between.

We are working on the the planar airpermeability measurement.

Best regards,

dr ir Lieven Anaf

Technology Manager

Bekaert Fibre Technologies

6 samples of each type

a)

Composition :

Bipolar plate stainless 0,4 mm

Sintering together : 2100 gr/m² 22 μm   2100 gr/m²
22 μm + bipolar plate

Thickness after sintering :   about   2,5 mm

Porosity 22 μm   about   89,5%
after pressing by Nuvera   1,65   mm
Porosity 22 μm   about   **84,10%**

b)

Composition :

Bipolar plate stainless 0,4 mm

1800 gr/m² 22 μm   1800 gr/m²

600 gr/m² 8 μm   600 gr/m²

8 μm was sintered before and pressed at 70 % porosity
Thickness after pressing 0,2 mm

Sintering together 8 + 22 μm + bipolar plate

Thickness after sintering 22 μm: about   2,4 mm

Porosity   22 μm   about   90,6%
after pressing by Nuvera   1,45   mm
Porosity 22 μm   about   **84,50%**

c)

<u>Composition :</u>

Bipolar plate stainless 0,4 mm

300 gr/m² 22 µm

1 wire mesh thickness 0,66 mm

600 gr/m² 22 µm

600 gr/m² 8 µm

8 µm was sintered before and pressed at 70 % porosity
Thickness after pressing 0,2 mm

Sintering together 8 + 22 µm + wire mesh + 22 µm + bipolar plate

Thickness after sintering about :

8 µm	0,2	mm
22 µm	0,71	mm
Wire mesh	0,66	mm
22 µm	1,43	mm

Porosity 22 µm	about	94,7%	
after pressing by Nuvera		0,79	mm
Porosity 22 µm	about	**85,80%**	

EXHIBIT C

Dossier: 5583

Titel: ~~bipolar~~ plate *(handwritten: tussen metal fuel cell on electrode plaatjes)*

5 Aanvrager: NV BEKAERT

Uitvinders: Anaf

x

x

10 Losfeld

x

x

15 x

x

x

Figuur abstract: figuur x

20

10/19/2001

XXXX ep aanvrage draft bipolar cell 1.doc

(handwritten: for cell in ... elec. [diagram] / STACK (struck: border) < fuel cell.)

Field of the Invention.

The present invention relates to bipolar plates, to be used in fuel cells or electrolysers,

5

Background of the invention.

(handwritten: PEM & electrolyser stacks)

Bipolar plates are generally known in the art. Such bipolar plates usually have at least 3 layers, engaging each other closely.

10

A first layer is a water- and gas-proof layer, hereafter referred to as "collector layer" This collector layer avoids gas or water leakage from one cell to an other, and guides electrons (e⁻) to or from the cell. Therefor it is generally known to use a ~~metal foil or plate~~.

(handwritten: or bipolar plate 15)

(handwritten: Conductive plate, most usually graphite plate.)

A second layer, engaging closely one side of the collector layer, is used to distribute the gasses used or provided by the chemical reaction in the fuel cell or electrolyser at the proton exchange membrane (PEM), over the whole surface of the fuel cell or electrolyser. This layer is hereafter

20 referred to as ~~"distribution layer"~~.

(handwritten: gas diffusion layer.)

A third layer, engaging closely the other side of the collector layer, is a layer, used to provide the contact between diffusion layer and PEM. At this so-called contact layer or electrode layer, the chemical reaction

25 takes place, due to the presence of catalytic elements, either on the contact layer or the PEM itself. Gasses, being provided via the diffusion layer to this contact layer, are to be retained sufficiently to enable the chemical reaction to take place.

30 The contact layer, and possibly also the diffusion layer, may be made hydrophobic, e.g. by impregnation or presence of hydrophobic elements such as Teflon®.

Depending on the place of the bipolar plate in the fuel cell or the electrolyser, a chemical reaction takes place in which e-, protons (H+) and a gas is consumed or provided near a PEM.

5   The H+ are provided or evacuated via the PEM to the chemical reaction. therefor, the contact between contact layer and PEM is to be as perfect as possible, since the chemical reaction preferably takes place as close as possible near the surface of the PEM.

10   The e- are provided or evacuated via the stack of collector layer, diffusion layer and contact layer. Therefor, all layers engaged are to be electro-conductive, and the resistance over the stack, and especially the contact resistance at the contacts of the several layers is to be as low as possible.

15

The diffusion layer is to spread the gas flow as much as possible over the whole surface of the contact layer, in order to use the present catalytic elements as complete as possible, providing e- and H+ over the whole surface of the PEM.

20   An example of a bipolar plate is described in XXXXX LYNTECH XXXXX.

Summary of the invention.

25   The present invention has as an object to provide an improved bipolar plate for use in fuel cells or electrolysers, which has a reduced electrical resistance over its thickness, an improved property of diffusing the reaction gasses over the surface of the PEM, against which it is to be used, and an improved contact between bipolar plate and PEM.

30

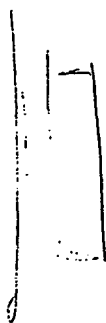A bipolar plate as subject of the invention comprises at least three layers:

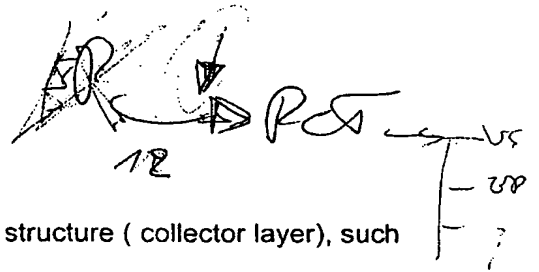However, the porosity of the contact layer is kept less than 80%

mesh ode in 1st loop.

⮑ nodes: laagje vkd los
mesh x plaat

- a gas- and liquid impermeable metal structure (collector layer), such as a metal foil or metal plate;

- A first metal fiber layer (diffusion layer), comprising metal fibers;

- A second metal fibers layer (contact layer), comprising metal fibers.

5

According to the present invention, the impermeable structure is sintered to one side of the diffusion layer, and the second metal fiber layer is sintered to the other side of the diffusion layer. The term layer is to be understood as an essentially flat object having a thickness which is

10 essentially equal over the surface of the object. The properties of both metal fiber layers are chosen in such a way that the planar air permeability is more than XXXXX, whereas the perpendicular air permeability of the second metal fibers layer is less than XXXXX.

15 Another bipolar plate as subject of the invention comprises

- a gas- and liquid impermeable metal structure (collector layer), such as a metal foil or metal plate;

- two first metal fiber layers (diffusion layers), comprising metal fibers;

- two second metal fiber layers (contact layers), comprising metal

20 fibers

It is understood that at both sides of the impermeable metal structure, a first metal fiber layer is sintered, and that a second metal fibers layer is sintered to each of the sides of the first metal fibers layers, which are not

25 connected to the impermeable metal structure. The planar air permeability at both sides of the impermeable metal structure is more than XXXXX, whereas the perpendicular air permeability of the second metal fibers layers are less than XXXXX.

30 With planar air permeability is meant the amount of gas which is passed through the metal fibers layer in a direction parallel to the plane of the

(A)

(4) Preferably, the AP ⊥ of the contact layer is less than xxx most preferably less than xxx

(**)

Preferably the porosity of the diffusion layer is more than 10% larger of the porosity of the contact layer most preferably, the porosity of the diffusion layer is more than

(3)

$$80\%$$
$$82\%$$
$$85\%$$
$$90\%$$

In order to obtain a preferred system plate AD, the thickness of the contact diffusion layer is more than xx, most preferably more than xxx. However, a too thick diffusion layer is to be avoided in order to keep the weight in reasonable range. It is clear that the higher the porosity of the diffusion layer, the thicker this layer may be chosen, keeping the same weight specifications

                                                      less
The thickness of the diffusion layer is preferably kept lower than xxx, most preferable lower than xxx.
                        less
This since the function of this layer is to VERTRAGEN from the reaction gasses contacting the BPM. A too thick layer, more than xx, does not provide any improvement.

(B)   The ep. ⌀ of the metal fibers used to provide the
diffusion layer, is preferably more than ...

$$12\,\mu$$
$$90\,\mu$$
$$22\,\mu$$
$$35\,\mu$$

meet methode

6

$\rightarrow$ // AP van totaal PCM zyde $>$ ...

$\hookrightarrow$ interst. gedefinieerd door diffusion-layer

$\hookrightarrow$ wyze van meten + plaats : midden byter cell. vte

$\oplus$

$\rightarrow$ diffuselaag: parallel $< 8\%$

(= dunne laag)

Teplest, ooka $\phi 20 \text{ cm}^2$.

$\wedge$ rel $< 75\%$

$< 72,5\%$

$\Rightarrow$ Daana nudclaus   - AP $\perp$ onderlaag $<$ ...

$\underline{\text{verk } \phi}$ = parallel verdeellaag $> 80\%$

perfectly $+$ 10 % > onderlaag

$> 80\%$

$> 82\%$

$> 85\%$

$> 8.\%$

- verel ϕ : boverlaag $> 12\mu$ $> 20\mu$ $> 22\mu$ $> 35\mu$.

onderlaag $< 12\mu$ $< 10\mu$ $< 8\mu$.

- aental draadnet in boverlaag.

- dikke lagen   boverlaag $>$ ...

onderlaag $<$ ...

metal SS - Ni - Ti

metal gas
barrier

Porous metal flow
field
  ↳ foam
  ↳ expanded sheet
  ↳ sintered particles
  ↳ sintered fibers
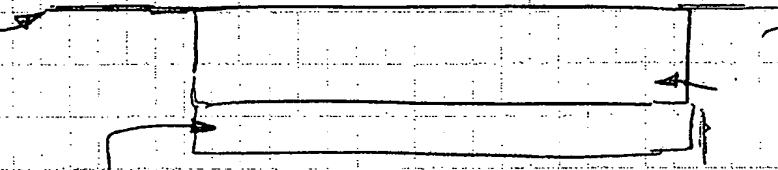
gas diffusion layer

  ↳ sintered particles
  ↳ sintered fibers

on either door welding
brazing
soldering
sintering
fusion bonding
vacuum bonding
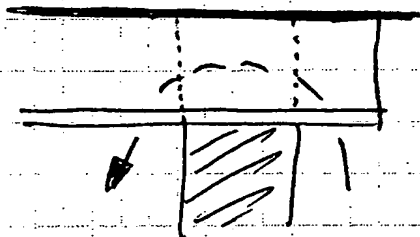
layer, measured XXXXX beschrijving van meetwijze. The planar air permeability is expressed in XXXXXXX eenheid.

With perpendicular air permeability is meant the amount of gas which is passed through the metal fibers layer in a direction perpendicular to the plane of the layer, measured XXXXX beschrijving van meetwijze. The planar air permeability is expressed in XXXXXXX eenheid.

Preferably the impermeable metal structure is a metal foil or metal plate, most preferably provided out of stainless steel, Ni or Ti.

The metal fibers used to provide the metal fibers layers are preferably stainless steel fibers, Ni-fibers or Ti-fibers. The metal fibers may be obtained by using presently known techniques, such as bundle drawing, coil shaving or any other production technique.

The equivalent diameter of the metal fibers used to provide the first metal fiber layer or layers, so-called diffusion layer, is larger than XXXXX, most preferably larger than XXXX. Possibly, more than one sheet of metal fibers is used to provide the first metal fibers layer or layers.

The density of the diffusion layers is preferably less than XXXXXX, whereas the thickness of the diffusion layers is preferably at least XXXXXX. Such diffusion layers provide to the largest extend the planar air permeability to the bipolar plate, and hence a superior distribution of the reaction gas over the whole surface of the adjacent contact layer or layers.

With "equivalent diameter of a metal fiber" is meant the diameter of an imaginary circle which has the same surface as a cross-section of the metal fiber.

-5-

With "density" in meant the ratio of the weight per volume of the layer over the weight of the same volume, provided 100% out of the metal alloy out of which the metal fibers are provided. The density is expressed in percentage. The "porosity" of a layer is expressed as

Porosity=100 - Density.

Alternatively, but also according to the present invention, a metal mesh may be inserted between two layers of metal fibers, comprised in this first metal fibers layer.

The equivalent diameter of the metal fibers used to provide the second metal fibers layer, so-called contact layer, is smaller than XXXXX, most preferably smaller than XXXX. Possibly, more than one sheet of metal fibers is used to provide the second metal fibers layer.
The density of the contact layer is preferably more than XXXXX, whereas the thickness of the contact layer is preferably less than XXXXXX.

Such contact layers, obtaining reaction gas via the diffusion layer over its whole surface, retains the gas sufficiently to propagate the chemical reaction at its reactive side, being the side of the contact layer, which contacts the adjacent PEM. Since the perpendicular air permeability of the second fm layer is relatively low, a too quick consumption of reaction gas at the gas entrance of the bipolar plate is prevented, which results in a presence of reactive gas over the whole surface of the connection layer. Possibly, the side of the contact layer, which contacts the PEM, is provided with an appropriate catalyst. Alternatively the catalyst is present on the surface of the PEM. Due to the small equivalent diameter of the metal fibers used to provide the contact layer, and the density of this layer, a very high degree of contact between contact layer and PEM may be obtained, whereas the side of the layer contacting the PEM, is relatively soft. Metal fibers projecting out of the essentially flat surface of

the contact layer, will not penetrate through the PEM when used, but will bend to the contact layer surface during assembly and use of the fuel cell or electrolyser. XXXXX kenmerk van zachtheid ???XXXXX

[handwritten margin notes and symbols]

5   Further, the obtained bipolar plate as subject of the invention has a transversal electrical resistance of less than XXXXXX Ohm in case a stack of an impermeable metal structure, a diffusion layer and a contact layer is provided. With transversal resistance is meant the electrical resistance measured between a point on the surface of the impermeable

10   metal structure and the point of the side of the connecting layer to be used against the PEM, which point is closest to the point of the impermeable metal structure. This low resistance is obtained thanks to the many contact points between several fibers being sintered to each other. *[handwritten: at to the collector layer (bipolar plate)]*

15   Alternatively, when at each side of the impermeable metal structure a diffusion layer and a connection layer is provided, the transversal electric resistance is less than XXXX Ohm.

It is clear that all three layers may be sintered to each other during one

20   sintering operation, either batch or continuous sintering, or that the diffusing and connecting layer are first sintered individually, before they are stacked and sintered to the impermeable metal structure.

*[handwritten margin: diffuser / poeder ofte outerin]*

25   Possibly, but not necessarily, the metal fiber layers may be impregnated with a water-repellent agent, e.g. polytertrafluoethylene such as Teflon®.

*[handwritten: or water — hydrofluoric of hydrofobic / e.p. / → Al₂O₃]*

XXXXXXXXXXXXXXX

Waarom is het niet voor de hand liggend dat de AIR PERMEABILITY en

30   de andere kenmerken specifiek gekozen moeten worden????

xXXXXXXXXXXXXXXXXX
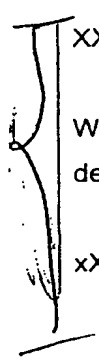
Such bipolar plates may be used in fuel cell, where at least two bipolar plates as subject of the invention are used. Between a contact layer of the first bipolar plate and a contact layer of a second bipolar plate, a

5   PEM is provided. At both sides of the PEM, necessary catalysts are present to propagate the chemical reaction wanted. To the diffusion layer of the first bipolar plate, H2 is provided, which flows through the whole diffusion layer (due to the relatively low planar air permeability of the bipolar plate). At the PEM, a reaction as underneath is propagated:
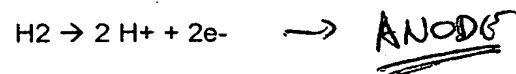
10

$$H2 \rightarrow 2 H+ + 2e- \quad \longrightarrow \quad \text{ANODE}$$

H+ is conducted through the PEM to the opposite side of the PEM, whereas the e- is drained away through the electrically conductive

15   connection and diffusion layer to the impermeable metal structure.

The e- is guided via an electrical circuit to the other impermeable metal structure of the second bipolar plate. Again via the diffusion and connection layer of this second bipolar plate, the e- are provided to the

20   chemical reaction at this side of the PEM, *see p Cathode*

O2 is provided to the diffusion layer of this second bipolar plate, which is conducted through the connection layer to the surface of the PEM. Here a reaction takes place, using O2, e- and H+ (provided through the PEM):

25

$$O2 + 4H+ + 4e- \rightarrow 2H2O$$
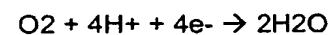
Since the optimal planar air permeability of the bipolar plate, and the relatively low perpendicular air permeability of the connection layers, the

30   gasses are spread in an optimal way over the whole PEM surface. Further, since the low electrical transversal resistance of the bipolar

plates, the e- are conducted to the electrical circuit without a significant
loss of power. *(due to joule-effect .*

where our joule-effect).

A similar benefit is made when the bipolar plates are used in
electrolysers. An identical combination of at least two bipolar plates is
provided. A determined tension is provided between the two
impermeable metal structures. At the bipolar plate with a positive
tension on its impermeable metal structure, H2O is provided, which
reacts at the PEM surface as:
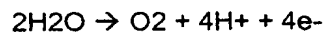
$$2H2O \rightarrow O2 + 4H+ + 4e-$$ *? ANODE ?*

the H+ is conducted through the PEM to the other side of the PEM,
whereas the e- is conducted via the metal fiber layers to the
impermeable metal structure. O2 is easily evacuated since the high
planar air permeability of the diffusion layer.

At the other side, a reaction takes place:

$$2H+ + 2e- \rightarrow H2,$$ *a KATHODE ?*

Where H+ is provided via the PEM and the e- are provided via the
impermeable metal structure (being negative pole) and the metal fiber
layers. H2 is easily evacuated due to the high planar air permeability of
the diffusion layer.

Brief description of the drawings.

The invention will now be described into more detail with reference to the accompanying drawings wherein

-FIGURE 1 x.

5   -

Description of the preferred embodiments of the invention.

xx

CLAIMS

1. A bipolar plate comprising an impermeable metal structure, at least one first metal fiber layer and at least one second metal fiber layer, said impermeable metal structure is sintered to one side of the first metal fibers layer, said second metal fibers layer is sintered to the other side of said first metal fibers layer, characterized in that the planar air permeability of the bipolar plate is more than XXXXX, the perpendicular air permeability of said second metal fibers layer is less than XXXXX.

2. A bipolar plate comprising an impermeable metal structure, two first metal fiber layers and two second metal fiber layers, said first metal fiber layers are sintered each to one side of the impermeable metal structure, said second metal fiber layers are sintered to the other sides of said first metal fiber layers characterized in that the planar air permeability of the bipolar plate at both sides of said impermeable metal structure is more than XXXXX, the perpendicular air permeability of said second metal fiber layers is less than XXXXX.

3. A bipolar plate as in claim 1 or 2, said first metal fiber layers having a density of less than xxxxxx

4. A bipolar plate as in claim 1 to 3, said second metal fiber layers having a density of more than xxxxxx

5. A bipolar plate as in claim 1 to 4, said first metal fiber layers comprising fibers with equivalent diameter of more than xxxx

6. A bipolar plate as in claim 1 to 5, said second metal fibers layers comprising fibers with equivalent diameter of less than xxxx

7. A bipolar plate as in claim 1 to 6, said first metal fiber layers having a thickness of more than xxxxx

8. A bipolar plate as in claim 1 to 7, said second metal fiber layers having a thickness of less than xxxxx

9. A bipolar plate as in claim 1 to 8, said bipolar plate having a transversal electric resistance less than XXXX Ohm

10. A bipolar plate as in claim 1 to 9, said impermeable metal structure being a metal plate.

11. A bipolar plate as in claim 1 to 9, said impermeable metal structure being a metal foil.

12. A bipolar plate as in claim 1 to 11, said metal fibers being stainless steel fibers.

13. A bipolar plate as in claim 1 to 11, said metal fibers being Ni-fibers

14. A bipolar plate as in claim 1 to 11, said metal fibers being Ti-fibers

15. A bipolar plate as in claim 1 to 14, said metal fibers having the same alloy of said impermeable metal structure.

16. A fuel cell, comprising bipolar plates as in claim 1 to 15.

17. An electrolyser, comprising bipolar plates as in claim 1 to 15.

18. The use of a bipolar plate as in claim 1 to 15 in fuel cells or electrolysers.

ABSTRACT

X.

EXHIBIT D

Dossier:	5583
Titel:	porous metal stack for fuel cells or electrolysers.
Aanvrager:	NV BEKAERT
Uitvinders:	Anaf
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	Losfelt
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Figuur abstract:	figuur x

11/8/2001

5583 ep aanvrage draft bipolar cell 2.doc

Field of the invention.

The present invention relates to a porous stack comprising metal fibers, to be used in fuel cells or electrolysers.

Background of the invention.

Fuel cells and electrolysers usually comprise a number of stacks, which are added in combination with proton exchange membrane (PEM), in order to obtain separated cells for the electrochemical reactions in either the fuel cell or electrolyser. Such stacks are generally known in the art. They usually have at least 3 layers, engaging each other closely.

A first layer is a water- and gas-proof layer, hereafter referred to as "collector layer", and also referred to in the art as "bipolar plate". This collector layer avoids gas or water leakage from one cell to an other, and guides electrons (e⁻) to or from the cell. Therefor it is generally known to use a conductive plate, usually a graphite plate.

A second layer, engaging closely one side of the collector layer, is used to distribute the gasses, used or provided by the electrochemical reaction in the fuel cell or electrolyser at the proton exchange membrane (PEM), over the whole surface of the fuel cell or electrolyser. This layer is hereafter referred to as "distribution layer".

A third layer, engaging closely the other side of the collector layer, is a layer, used to provide the contact between diffusion layer and PEM. At this so-called contact layer or "electrode layer", the electrochemical reaction takes place, due to the presence of catalytic elements, either on the contact layer or the PEM itself. Gasses, being provided via the diffusion layer to this contact layer, are to be retained sufficiently to enable the electrochemical reaction to take place.

The contact layer, and possibly also the diffusion layer, may be made hydrophobic, e.g. by impregnation or presence of hydrophobic elements such as Teflon®, or hydrophilic.

5   Depending on the place of the stack in the fuel cell or the electrolyser, a electrochemical reaction takes place in which e-, protons (H+) and a gas are consumed or provided near a PEM.

The H+ are provided or evacuated via the PEM to the electrochemical
10   reaction. Therefor, the contact between contact layer and PEM is to be as perfect as possible, since the electrochemical reaction takes place at the catalytic layer, which is close to the surface of the PEM.

The e- are provided or evacuated via the stack of collector layer,
15   diffusion layer and contact layer. Therefor, all layers engaged are to be electro-conductive, and the resistance over the stack, and especially the contact resistance at the contacts of the several layers is to be as low as possible.

20   The diffusion layer is to spread the gas flow as much as possible over the whole surface of the contact layer, in order to use the present catalytic elements as complete as possible, providing e- and H+ over the whole surface of the PEM.

25   An example of a stack is described in WO0069003 and EP0141241.

Summary of the invention.

The present invention has as an object to provide an improved stack for
30   use in fuel cells or electrolysers, which comprises metal fibers and which has a reduced electrical resistance over its thickness, an improved property of diffusing the reaction gasses over the surface of the PEM,

against which it is to be used, and an improved contact between stack and PEM.

A stack as subject of the invention comprises at least three layers:

5
- a gas- and liquid impermeable metal structure (collector layer), such as a metal foil or metal plate;
- A first metal fiber layer (diffusion layer), comprising metal fibers;
- A second metal fibers layer (contact layer), comprising metal fibers.

10
According to the present invention, the impermeable structure is sintered to one side of the diffusion layer, and the second metal fiber layer is sintered to the other side of the diffusion layer. The term layer is to be understood as an essentially flat object having a thickness, which is essentially equal over the surface of the object. The properties of both

15
metal fiber layers are chosen in such a way that the planar air permeability of the stack as subject of the invention is more than XXXXX, whereas the porosity of the contact layer is less than 80% or even less than 75%.

20
Another stack as subject of the invention comprises
- a gas- and liquid impermeable metal structure (collector layer), such as a metal foil or metal plate;
- two first metal fiber layers (diffusion layers), comprising metal fibers;
- two second metal fiber layers (contact layers), comprising metal

25
fibers

It is understood that at both sides of the impermeable metal structure, a first metal fiber layer is sintered, and that a second metal fibers layer is sintered to each of the sides of the first metal fibers layers, which are not

30
connected to the impermeable metal structure. The planar air permeability of the stack as subject of the invention at both sides of the

impermeable metal structure is more than XXXXX, whereas the porosity of the contact layers are less than 80% or even less than 75%.

With planar air permeability is meant the amount of gas which is passed through the metal fibers layers of the stack in a direction parallel to the plane of the layers, measured XXXXX beschrijving van meetwijze. The planar air permeability is expressed in XXXXXXX eenheid.

The "porosity" of a layer is expressed as

Porosity = 100 - Density.

With "density" in meant the ratio of the weight per volume of the layer over the weight of the same volume, provided 100% out of the metal alloy out of which the metal fibers are provided. The density is expressed in percentage.

Preferably the impermeable metal structure is a metal foil or metal plate, most preferably provided out of stainless steel, Ni or Ti. In case of stainless steel, preferably Fe-Ni-Cr alloys are used such as alloys of the serie AISI-300 and AISI-400, preferably AISI 316L or Ni-Fe alloys such as Inconel®.

The metal fibers used to provide the metal fibers layers are preferably stainless steel fibers, Ni-fibers or Ti-fibers. In case of stainless steel fibers, preferably Fe-Ni-Cr alloys are used such as alloys of the serie AISI-300 and AISI-400, preferably AISI 316L or Ni-Fe alloys such as Inconel®. The metal fibers may be obtained by using presently known techniques, such as bundle drawing, coil shaving or any other production technique.

Preferably all layers of the stack are provided out of the same metal or metal alloy.

The contact layer and the diffusion layer may be sintered separately, before stack comprising a collector layer, one or more diffusion layers and one or more contact layers are assembled and sintered to each other. Alternatively, the diffusion layers and the contact layers, both comprising metal fibers, and the collector layer may be sintered to each other all at once during one sintering operation.

The equivalent diameter of the metal fibers used to provide the first metal fiber layer or layers, so-called diffusion layer, is larger than 20µm, most preferably larger than 50µm. Possibly, more than one sheet of metal fibers is used to provide the first metal fibers layer or layers.

With "equivalent diameter of a metal fiber" is meant the diameter of an imaginary circle which has the same surface as a cross-section of the metal fiber.

Preferably, the porosity of the diffusion layer is more than 10% larger than the porosity of the contact layer. Most preferably, the porosity of the diffusion layer is more than 80%, or even more than 82%, such as more than 85% or even more than 90%. In order to obtain a preferred stack as subject of the invention the thickness of the diffusion layer is more than 0.5mm, most preferably more than 1mm.
Such diffusion layers provide to the largest extend the planar air permeability to the stack, and hence a superior distribution of the reaction gas over the whole surface of the adjacent contact layer or layers.

Alternatively, but also according to the present invention, a metal mesh or stretch metal foil or plate may be inserted between two layers of metal fibers, comprised in this first metal fibers layer.

5   The equivalent diameter of the metal fibers used to provide the second metal fibers layer, so-called contact layer, is smaller than 30μm, most preferably smaller than 10μm. Possibly, more than one sheet of metal fibers is used to provide the second metal fibers layer.
The thickness of the contact layer is preferably less than 0.2mm.

10   Such contact layers, obtaining reaction gas via the diffusion layer over its whole surface, retains the gas sufficiently to propagate the electrochemical reaction at its reactive side, being the side of the contact layer, which contacts the adjacent PEM. Since the perpendicular air permeability of the second metal fiber layer is relatively low, a too quick

15   consumption of reaction gas at the gas entrance of the stack is prevented, which results in a presence of reactive gas over the whole surface of the connection layer. Possibly, the side of the contact layer, which contacts the PEM, is provided with an appropriate catalyst. Alternatively the catalyst is present on the surface of the PEM. Due to

20   the small equivalent diameter of the metal fibers used to provide the contact layer, and the density of this layer, a very high degree of contact between contact layer and PEM may be obtained, whereas the side of the layer contacting the PEM, is relatively soft. Metal fibers projecting out of the essentially flat surface of the contact layer, will not penetrate

25   through the PEM when used, but will bend to the contact layer surface during assembly and use of the fuel cell or electrolyser. XXXXX kenmerk van zachtheid ???XXXXX

Preferably, the perpendicular air permeability of the contact layer is less

30   than xxxxxxx, most preferably even less than xxxxx.
With perpendicular air permeability is meant the amount of gas which is passed through the metal fibers layer in a direction perpendicular to the

plane of the layer, measured XXXXX beschrijving van meetwijze. The planar air permeability is expressed in XXXXXXX eenheid.

Further, the obtained stack as subject of the invention has a transversal electrical resistance of less than XXXXXX Ohm in case a stack of an impermeable metal structure, a diffusion layer and a contact layer is provided. With transversal resistance is meant the electrical resistance measured between a point on the surface of the impermeable metal structure and the point of the side of the connecting layer to be used against the PEM, which point is closest to the point of the impermeable metal structure. This low resistance is obtained thanks to the many contact points between several fibers being sintered to each other or to the collector layer.

Alternatively, when at each side of the impermeable metal structure a diffusion layer and a connection layer is provided, the transversal electric resistance is less than XXXX Ohm.

It is clear that all three layers may be sintered to each other during one sintering operation, either batch or continuous sintering, or that the diffusion and connecting layer are first sintered individually, before they are stacked and sintered to the impermeable metal structure.

After sintering, possibly, but not necessarily, the metal fiber layers may be impregnated with a hydrophobic or hydrophilic agent, e.g. polytertrafluoethylene such as Teflon® as hydrophobic agent.
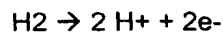
XXXXXXXXXXXXXXX

Waarom is het niet voor de hand liggend dat de AIR PERMEABILITY en de andere kenmerken specifiek gekozen moeten worden????

xXXXXXXXXXXXXXXXXXX

Such stacks may be used in fuel cell, where at least two stacks as subject of the invention are used. Between a contact layer of the first stack and a contact layer of a second stack, a PEM is provided. At both sides of the PEM, necessary catalysts are present to propagate the electrochemical reaction wanted. To the diffusion layer of the first stack, H2 is provided, which flows through the whole diffusion layer (due to the relatively low planar air permeability of the stack). At the PEM, a reaction as underneath is propagated:

$$H2 \rightarrow 2 H+ + 2e-$$

This side of the fuel cell is referred to as anode.
H+ is conducted through the PEM to the opposite side of the PEM, whereas the e- is drained away through the electrically conductive connection and diffusion layer to the impermeable metal structure.

The e- is guided via an electrical circuit to the other impermeable metal structure of the second stack. Again via the diffusion and connection layer of this second stack, the e- are provided to the electrochemical reaction at this side of the PEM, being the cathode side.
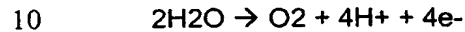
O2 is provided to the diffusion layer of this second stack, which is conducted through the connection layer to the surface of the PEM. Here a reaction takes place, using O2, e- and H+ (provided through the PEM):

$$O2 + 4H+ + 4e- \rightarrow 2H2O$$

Since the optimal planar air permeability of the stack, and the relatively low perpendicular air permeability of the connection layers, the gasses are spread in an optimal way over the whole PEM surface. Further, since

the low electrical transversal resistance of the stacks, the e- are conducted to the electrical circuit without a significant loss of power.

A similar benefit is made when the stacks are used in electrolysers. An identical combination of at least two stacks is provided. A determined tension is provided between the two impermeable metal structures. At the stack with a positive tension on its impermeable metal structure, H2O is provided, which reacts at the PEM surface as:
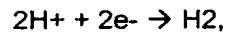
$$2H2O \rightarrow O2 + 4H+ + 4e-$$

xxx anode ????xxx

the H+ is conducted through the PEM to the other side of the PEM, whereas the e- is conducted via the metal fiber layers to the impermeable metal structure. O2 is easily evacuated since the high planar air permeability of the diffusion layer.

At the other side, a reaction takes place:

$$2H+ + 2e- \rightarrow H2,$$

??? cathode ????xxx

Where H+ is provided via the PEM and the e- are provided via the impermeable metal structure (being negative pole) and the metal fiber layers. H2 is easily evacuated due to the high planar air permeability of the diffusion layer.

5 **Brief description of the drawings.**

The invention will now be described into more detail with reference to the accompanying drawings wherein

-FIGURE 1 x.

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Description of the preferred embodiments of the invention.

xx

CLAIMS

1. A stack comprising an impermeable metal structure, at least one first metal fiber layer and at least one second metal fiber layer, said impermeable metal structure is sintered to one side of the first metal fibers layer, said second metal fibers layer is sintered to the other side of said first metal fibers layer, characterized in that the planar air permeability of the stack is more than XXXXX, porosity of said second metal fiber layer is less than 80%.

2. A stack comprising an impermeable metal structure, two first metal fiber layers and two second metal fiber layers, said first metal fiber layers are sintered each to one side of the impermeable metal structure, said second metal fiber layers are sintered to the other sides of said first metal fiber layers characterized in that the planar air permeability of the stack at both sides of said impermeable metal structure is more than XXXXX, porosity of said second metal fiber layers is less than 80%..

3. A stack as in claim 1 or 2, said first metal fiber layers having a porosity of more than 80%

4. A stack as in claim 1 to 3, said second metal fiber layers having a planar air permeability of less than XXXX

5. A stack as in claim 1 to 4, said first metal fiber layers comprising fibers with equivalent diameter of more than 20µm

6. A stack as in claim 1 to 5, said second metal fibers layers comprising fibers with equivalent diameter of less than 30µm

7. A stack as in claim 1 to 6, said first metal fiber layers having a thickness of more than 0.5mm

8. A stack as in claim 1 to 7, said second metal fiber layers having a thickness of less than 0.2mm

9. A stack as in claim 1 to 8, said stack having a transversal electric resistance less than XXXX Ohm

10. A stack as in claim 1 to 9, said impermeable metal structure being a metal plate.

11. A stack as in claim 1 to 9, said impermeable metal structure being a metal foil.

12. A stack as in claim 1 to 11, said metal fibers being stainless steel fibers.

13. A stack as in claim 1 to 11, said metal fibers being Ni-fibers

14. A stack as in claim 1 to 11, said metal fibers being Ti-fibers

15. A stack as in claim 1 to 14, said metal fibers having the same alloy of said impermeable metal structure.

16. A fuel cell, comprising stacks as in claim 1 to 15.

17. An electrolyser, comprising stacks as in claim 1 to 15.

18. The use of a stack as in claim 1 to 15 in fuel cells or electrolysers.

ABSTRACT

X.

EXHIBIT E

ASO = An subject of the invention
F = Figure

An embodiment of a porous stack an ASO is shown in Figure 1.

The stack ^{10} comprises a gas- and liquid unpermeable metal structure, so-called collector layer 11. In the embodiment shown in F1, a metal plate, provided out of stainless steel with alloy AISI 316L is used. The collector layer has a thickness of 0,4 MM.

SINTERED

To one side of this collector layer, a diffusion layer 12, being a metal fiber layer consisting out of stainless steel fibers (Alloy AISI 316 L) with equivalent diameter of 22 μm is provided. This diffusion layer 12 has a thickness of 1.7 mm and has a porosity of 85%.

The fibers are obtained by coil-shaving process, which provides to the fibers a substantially rectangular cross-section, having a long side of approximately 25 μm and a short side of approximately 15 μm.

To the side of the diffusion layer, which does not contact the collector layer, a second metal fiber layer 13, being the contact layer, is provided.

This contact layer 13 consists of SINTERED bundle drawn stainless steel fibers (Alloy AISI 316 L) having an essentially circular cross-section with an equivalent diameter of 8 μm.

①

This contact layer 13 has a thickness of approximately 0,2 mm and a porosity of preferably 70%. Alternatively a porosity of 85% may be used.

An alternative stack 20 ASo is shown in F2. This embodiment comprises a collector layer 21, being identical to the collector layer of F1.

At both sides of the collector layer 21, a diffusion layer 22a and 22b is provided, each of diffusion layer 22a and 22b being identical as the diffusion layer of F1

Similar as in F1, a contact layer 23a and 23b is provided to the sides of the diffusion layer 22a a 22b. These contact layers 23a and 23b are identical as the contact layer of F1

The planar air permeability of stack 10, and of both sides 24a and 24b of stack 20, are measured as shown in F3.

*** NOG VERDER UIT TE WERKEN ***.

②

The planar air permeability of stack 10 or of both sides 24a and 24b of stack 10, are XXXX.

It was found that the porosity of contact layers 13, 23a or 23b does not make significant changes to this planar air permeability.

The perpendicular air permeability of the ~~and~~ contact layer 13, 23a and 23b are

Porosity	Perpendicular air permeability
70%	xxx
85%	xxx

For a thickness

Porosity	thickness	perpendicular air permeability
70%	xx	xx
85%	xx	xx

An electrical resistance may be measured over the stack 10 or over both sides of stack 10, being
*** NOG VERDER UITWERKEN ***

③

The stacks 10 or 20 Also may be used in an electrolyser or in a fuel cell, as shown in F4 and F5.

F4 shows a fuel cell 40, comprising serial stacks 10 and 20, separated from each other using proton exchange membranes 41, ~~which~~ between the contact layers of the stacks 10 or 20 and the PEM 41, appropriate catalysts are provided.

O_2 or H_2 is provided to the stack in such a way that at both sides of the PEM, an electrochemical reaction takes place. The e^- are collected through the contact and diffusion layers by the collector layers.

The collector layers are connected to each other via an appropriate ~~the~~ electrical connection device 42, which provides electrical current to & used by an electrical device or to a battery 43

In F5, two stacks 10 are separated from each other by means of a catalytically coated PEM 51.

by a
tension
~~the~~
source

An electrical tension is provided to the collector layers of the electrolyser. H_2O, being provided to the electrolyser, is ~~electrochemically~~ reacts electrochemically, providing O_2 and H_2.

(4)

EXHIBIT F

Dossier: 5583

Titel: porous metal stack for fuel cells or electrolysers.

5   Aanvrager: NV BEKAERT

Uitvinders: Anaf

x

x

10

Losfelt

x

x

15   x

x

x

Figuur abstract: figuur x

20

11/22/2001

5583 ep aanvrage draft bipolar cell 4.doc

Field of the Invention.

The present invention relates to a porous stack comprising metal fibers, to be used in fuel cells or electrolysers.

Background of the Invention.

Fuel cells and electrolysers usually comprise a number of stacks, which are added in combination with proton exchange membrane (PEM), in order to obtain separated cells for the electrochemical reactions in either the fuel cell or electrolyser. Such stacks are generally known in the art. They usually have at least 3 layers, engaging each other closely.

A first layer is a water- and gas-proof layer, hereafter referred to as "collector layer", and also referred to in the art as "bipolar plate". This collector layer avoids gas or water leakage from one cell to an other, and guides electrons (e⁻) to or from the cell. Therefor it is generally known to use a conductive plate, usually a graphite plate.

A second layer, engaging closely one side of the collector layer, is used to distribute the gasses, used or provided by the electrochemical reaction in the fuel cell or electrolyser at the proton exchange membrane (PEM), over the whole surface of the fuel cell or electrolyser. This layer is hereafter referred to as "distribution layer".

A third layer, engaging closely the other side of the collector layer, is a layer, used to provide the contact between diffusion layer and PEM. At this so-called contact layer or "electrode layer", the electrochemical reaction takes place, due to the presence of catalytic elements, either on the contact layer or the PEM itself. Gasses, being provided via the diffusion layer to this contact layer, are to be retained sufficiently to enable the electrochemical reaction to take place.

The contact layer, and possibly also the diffusion layer, may be made hydrophobic, e.g. by impregnation or presence of hydrophobic elements such as Teflon®, or hydrophilic.

5   Depending on the place of the stack in the fuel cell or the electrolyser, a electrochemical reaction takes place in which e-, protons (H+) and a gas are consumed or provided near a PEM.

The H+ are provided or evacuated via the PEM to the electrochemical
10   reaction. Therefor, the contact between contact layer and PEM is to be as perfect as possible, since the electrochemical reaction takes place at the catalytic layer, which is close to the surface of the PEM.

The e- are provided or evacuated via the stack of collector layer,
15   diffusion layer and contact layer. Therefor, all layers engaged are to be electro-conductive, and the resistance over the stack, and especially the contact resistance at the contacts of the several layers is to be as low as possible.

20   The diffusion layer is to spread the gas flow as much as possible over the whole surface of the contact layer, in order to use the present catalytic elements as complete as possible, providing e- and H+ over the whole surface of the PEM.

25   An example of a stack is described in WO0069003 and EP0141241.

Summary of the Invention.

The present invention has as an object to provide an improved stack for
30   use in fuel cells or electrolysers, which comprises metal fibers and which has a reduced electrical resistance over its thickness, an improved property of diffusing the reaction gasses over the surface of the PEM,

against which it is to be used, and an improved contact between stack and PEM.

A stack as subject of the invention comprises at least three layers:

- a gas- and liquid impermeable metal structure (collector layer), such as a metal foil or metal plate;
- A first metal fiber layer (diffusion layer), comprising metal fibers;
- A second metal fibers layer (contact layer), comprising metal fibers.

According to the present invention, the impermeable structure is sintered to one side of the diffusion layer, and the second metal fiber layer is sintered to the other side of the diffusion layer. The term layer is to be understood as an essentially flat object having a thickness, which is essentially equal over the surface of the object. The properties of both metal fiber layers are chosen in such a way that the planar air permeability of the stack as subject of the invention is more than XXXXX, whereas the porosity of the contact layer is less than 80% or even less than 75%.

Another stack as subject of the invention comprises

- a gas- and liquid impermeable metal structure (collector layer), such as a metal foil or metal plate;
- two first metal fiber layers (diffusion layers), comprising metal fibers;
- two second metal fiber layers (contact layers), comprising metal fibers

It is understood that at both sides of the impermeable metal structure, a first metal fiber layer is sintered, and that a second metal fibers layer is sintered to each of the sides of the first metal fibers layers, which are not connected to the impermeable metal structure. The planar air permeability of the stack as subject of the invention at both sides of the

impermeable metal structure is more than XXXXX, whereas the porosity of the contact layers are less than 80% or even less than 75%.

With planar air permeability is meant the amount of gas which is passed through the metal fibers layers of the stack in a direction parallel to the plane of the layers, measured XXXXX beschrijving van meetwijze. The planar air permeability is expressed in XXXXXXX eenheid.

The "porosity" of a layer is expressed as

$$Porosity = 100 - Density.$$

With "density" in meant the ratio of the weight per volume of the layer over the weight of the same volume, provided 100% out of the metal alloy out of which the metal fibers are provided. The density is expressed in percentage.

Preferably the impermeable metal structure is a metal foil or metal plate, most preferably provided out of stainless steel, Ni or Ti. In case of stainless steel, preferably Fe-Ni-Cr alloys are used such as alloys of the serie AISI-300 and AISI-400, preferably AISI 316L or Ni-Fe alloys such as Inconel®.

The metal fibers used to provide the metal fibers layers are preferably stainless steel fibers, Ni-fibers or Ti-fibers. In case of stainless steel fibers, preferably Fe-Ni-Cr alloys are used such as alloys of the serie AISI-300 and AISI-400, preferably AISI 316L or Ni-Fe alloys such as Inconel®. The metal fibers may be obtained by using presently known techniques, such as bundle drawing, coil shaving or any other production technique.

Preferably all layers of the stack are provided out of the same metal or metal alloy.

The contact layer and the diffusion layer may be sintered separately, before stack comprising a collector layer, one or more diffusion layers and one or more contact layers are assembled and sintered to each other. Alternatively, the diffusion layers and the contact layers, both comprising metal fibers, and the collector layer may be sintered to each other all at once during one sintering operation.

The equivalent diameter of the metal fibers used to provide the first metal fiber layer or layers, so-called diffusion layer, is larger than 20µm, most preferably larger than 50µm. Possibly, more than one sheet of metal fibers is used to provide the first metal fibers layer or layers.

With "equivalent diameter of a metal fiber" is meant the diameter of an imaginary circle which has the same surface as a cross-section of the metal fiber.

Preferably, the porosity of the diffusion layer is more than 10% larger than the porosity of the contact layer. Most preferably, the porosity of the diffusion layer is more than 80%, or even more than 82%, such as more than 85% or even more than 90%. In order to obtain a preferred stack as subject of the invention the thickness of the diffusion layer is more than 0.5mm, most preferably more than 1mm.
Such diffusion layers provide to the largest extend the planar air permeability to the stack, and hence a superior distribution of the reaction gas over the whole surface of the adjacent contact layer or layers.

Alternatively, but also according to the present invention, a metal mesh or stretch metal foil or plate may be inserted between two layers of metal fibers, comprised in this first metal fibers layer.

5   The equivalent diameter of the metal fibers used to provide the second metal fibers layer, so-called contact layer, is smaller than 30μm, most preferably smaller than 10μm. Possibly, more than one sheet of metal fibers is used to provide the second metal fibers layer.

The thickness of the contact layer is preferably less than 0.2mm.

10   Such contact layers, obtaining reaction gas via the diffusion layer over its whole surface, retains the gas sufficiently to propagate the electrochemical reaction at its reactive side, being the side of the contact layer, which contacts the adjacent PEM. Since the perpendicular air permeability of the second metal fiber layer is relatively low, a too quick

15   consumption of reaction gas at the gas entrance of the stack is prevented, which results in a presence of reactive gas over the whole surface of the connection layer. Possibly, the side of the contact layer, which contacts the PEM, is provided with an appropriate catalyst. Alternatively the catalyst is present on the surface of the PEM. Due to

20   the small equivalent diameter of the metal fibers used to provide the contact layer, and the density of this layer, a very high degree of contact between contact layer and PEM may be obtained, whereas the side of the layer contacting the PEM, is relatively soft. Metal fibers projecting out of the essentially flat surface of the contact layer, will not penetrate

25   through the PEM when used, but will bend to the contact layer surface · during assembly and use of the fuel cell or electrolyser. XXXXX kenmerk van zachtheid ???XXXXX

30   Preferably, the perpendicular air permeability of the contact layer is less than xxxxxxx, most preferably even less than xxxxx.

With perpendicular air permeability is meant the amount of gas which is passed through the metal fibers layer in a direction perpendicular to the

plane of the layer, measured XXXXX beschrijving van meetwijze. The planar air permeability is expressed in XXXXXXX eenheid.

Further, the obtained stack as subject of the invention has a transversal electrical resistance of less than XXXXXX Ohm in case a stack of an impermeable metal structure, a diffusion layer and a contact layer is provided. With transversal resistance is meant the electrical resistance measured between a point on the surface of the impermeable metal structure and the point of the side of the connecting layer to be used against the PEM, which point is closest to the point of the impermeable metal structure. This low resistance is obtained thanks to the many contact points between several fibers being sintered to each other or to the collector layer.

Alternatively, when at each side of the impermeable metal structure a diffusion layer and a connection layer is provided, the transversal electric resistance is less than XXXX Ohm.

It is clear that all three layers may be sintered to each other during one sintering operation, either batch or continuous sintering, or that the diffusion and connecting layer are first sintered individually, before they are stacked and sintered to the impermeable metal structure.
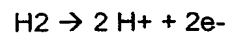
After sintering, possibly, but not necessarily, the metal fiber layers may be impregnated with a hydrophobic or hydrophilic agent, e.g. polytertrafluoethylene such as Teflon® as hydrophobic agent.

XXXXXXXXXXXXXX

Waarom is het niet voor de hand liggend dat de AIR PERMEABILITY en de andere kenmerken specifiek gekozen moeten worden????

xXXXXXXXXXXXXXXXXX

Such stacks may be used in fuel cell, where at least two stacks as subject of the invention are used. Between a contact layer of the first stack and a contact layer of a second stack, a PEM is provided. At both sides of the PEM, necessary catalysts are present to propagate the electrochemical reaction wanted. To the diffusion layer of the first stack, H2 is provided, which flows through the whole diffusion layer (due to the relatively low planar air permeability of the stack). At the PEM, a reaction as underneath is propagated:

$H2 \rightarrow 2 H+ + 2e-$

This side of the fuel cell is referred to as anode.
H+ is conducted through the PEM to the opposite side of the PEM, whereas the e- is drained away through the electrically conductive connection and diffusion layer to the impermeable metal structure.

The e- is guided via an electrical circuit to the other impermeable metal structure of the second stack. Again via the diffusion and connection layer of this second stack, the e- are provided to the electrochemical reaction at this side of the PEM, being the cathode side.
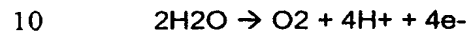
O2 is provided to the diffusion layer of this second stack, which is conducted through the connection layer to the surface of the PEM. Here a reaction takes place, using O2, e- and H+ (provided through the PEM):

$O2 + 4H+ + 4e- \rightarrow 2H2O$

Since the optimal planar air permeability of the stack, and the relatively low perpendicular air permeability of the connection layers, the gasses are spread in an optimal way over the whole PEM surface. Further, since

the low electrical transversal resistance of the stacks, the e- are
conducted to the electrical circuit without a significant loss of power.

A similar benefit is made when the stacks are used in electrolysers. An
identical combination of at least two stacks is provided. A determined
tension is provided between the two impermeable metal structures. At
the stack with a positive tension on its impermeable metal structure, H_2O
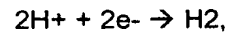is provided, which reacts at the PEM surface as:

$$2H_2O \rightarrow O_2 + 4H+ + 4e-$$

xxx anode ????xxx

the H+ is conducted through the PEM to the other side of the PEM,
whereas the e- is conducted via the metal fiber layers to the
impermeable metal structure. O_2 is easily evacuated since the high
planar air permeability of the diffusion layer.

At the other side, a reaction takes place:

$$2H+ + 2e- \rightarrow H_2,$$

?? cathode ???xxx

Where H+ is provided via the PEM and the e- are provided via the
impermeable metal structure (being negative pole) and the metal fiber
layers. H_2 is easily evacuated due to the high planar air permeability of
the diffusion layer.

5   **Brief description of the drawings.**

The invention will now be described into more detail with reference to the
accompanying drawings wherein
-FIGURE 1 x.

-

10

Description of the preferred embodiments of the invention.

An embodiment of a porous stack as subject of the invention is shown in
Figure 1.

15   The stack 10 comprises a gas- and liquid impermeable metal structure,
so-called collector layer 11. In the embodiment shown in Figure 1, a
metal plate provided out of stainless steel with alloy AISI 316L is used.
The collector layer has a thickness of 0,4 mm.

20   To one side of this collector layer, a diffusion layer 12, being a sintered
metal fiber layer consisting out of stainless steel fibers (Alloy AISI 316L)
with equivalent diameter of 22 µm is provided. This diffusion layer 12 has
a thickness of 1,7mm and has a porosity of 85%.

25   The fibers are obtained by coil shaving process, which provides to the
fibers a substantially rectangular cross-section, have a large side of
approximately 25 µm and a short side of approximately 15 µm

To the side of the diffusion layer, which does not contact the collector
30   layer, a second metal fiber layer 13, being the contact layer, is provided.

This contact layer 13 consists of sintered bundle drawn stainless steel fibers (Alloy AISI 136L) having an essentially circular cross-section with an equivalent diameter of 8 μm. This contact layer 13 has a thickness of approximately 0,2 mm and a porosity of preferably 70%. Alternatively a porosity of 85% may be used.

An alternative stack 20 as subject of the invention is shown in Figure 2. This embodiment comprises a collector layer 21, being identical to the collector layer of Figure 1.

At both sides of the collector layer 21, a diffusion layer 22a and 22b is provided, each diffusion layer 22a and 22b being identical as the diffusion layer of Figure 1.

Similar as in Figure 1, a contact layer 23a and 23b is provided to the sides of diffusion layer 22a and 22b. These contact layers 23a and 23b are identical as the contact layer of Figure 1.

The planar air permeability of stack 10, and of both sides 24a and 24b of stack 20, are measured as shown in Figure 3.
XXXXX nog verder uit te werken XXXXXXX

The planar air permeability of stack 10a of both sides 24a and 24b of stack 20 are XXXXX

It was found that the porosity of contact layer 13, 23a or 23b does not make significant changes to this planar air permeability.

The perpendicular air permeability of the contact layer 13, 23a and 23b are

Porosity	Thickness	Perpendicular air permeability
70%	XX	XX

85%	XX	XX

An electrical resistance may be measured over the stack 10 or over both sides of stack 20, being XXXXX nog verder uit te werken XXXXXX

5   The stacks 10 or 20 as subject of the invention may be used in an electrolyser or in a fuel cell, as shown in Figure 4 and Figure 5.

Figure 4 shows a fuel cell 40, comprising several stacks 10 and 20, separated from each other using proton exchange membranes 41,
10   between the contact layers of the stacks 10 or 20 and the PEM 41, appropriate catalysts are provided.

O_2 or H_2 is provided to the stack in such a way that at both sides of the PEM, an electrochemical reaction takes place. The e⁻ are collected
15   through the contact and diffusion layers by the collector layers.

The collector layers are connected to each other via an appropriate electrical connection device 42, which provided electrical current to be used by an electrical device or to a battery 43/

20
In Figure 5, two stacks 10 are separated from each other by means of a catolytically coated PEM 51.

An electrical tension is provided by a tension source to the collector
25   layers of the electrolyser. H_2O being provided to the electrolyser, reacts electrochemically, providing O_2 and H_2.

CLAIMS

1. A stack comprising an impermeable metal structure, at least one first metal fiber layer and at least one second metal fiber layer, said impermeable metal structure is sintered to one side of the first metal fibers layer, said second metal fibers layer is sintered to the other side of said first metal fibers layer, characterized in that the planar air permeability of the stack is more than XXXXX, porosity of said second metal fiber layer is less than 80%.

2. A stack comprising an impermeable metal structure, two first metal fiber layers and two second metal fiber layers, said first metal fiber layers are sintered each to one side of the impermeable metal structure, said second metal fiber layers are sintered to the other sides of said first metal fiber layers characterized in that the planar air permeability of the stack at both sides of said impermeable metal structure is more than XXXXX, porosity of said second metal fiber layers is less than 80%..

3. A stack as in claim 1 or 2, said first metal fiber layers having a porosity of more than 80%

4. A stack as in claim 1 to 3, said second metal fiber layers having a planar air permeability of less than XXXX

5. A stack as in claim 1 to 4, said first metal fiber layers comprising fibers with equivalent diameter of more than 20μm

6. A stack as in claim 1 to 5, said second metal fibers layers comprising fibers with equivalent diameter of less than 30μm

7. A stack as in claim 1 to 6, said first metal fiber layers having a thickness of more than 0.5mm

8. A stack as in claim 1 to 7, said second metal fiber layers having a thickness of less than 0.2mm

9. A stack as in claim 1 to 8, said stack having a transversal electric resistance less than XXXX Ohm

10. A stack as in claim 1 to 9, said impermeable metal structure being a metal plate.

11. A stack as in claim 1 to 9, said impermeable metal structure being a metal foil.

12. A stack as in claim 1 to 11, said metal fibers being stainless steel fibers.

13. A stack as in claim 1 to 11, said metal fibers being Ni-fibers

14. A stack as in claim 1 to 11, said metal fibers being Ti-fibers

15. A stack as in claim 1 to 14, said metal fibers having the same alloy of said impermeable metal structure.

16. A fuel cell, comprising stacks as in claim 1 to 15.

17. An electrolyser, comprising stacks as in claim 1 to 15.

18. The use of a stack as in claim 1 to 15 in fuel cells or electrolysers.

ABSTRACT

x.

EXHIBIT G

Field of the Invention.

The present invention relates to a porous stack comprising metal fibers, to be used in fuel cells or electrolysers.

5

Background of the Invention.

Fuel cells and electrolysers usually comprise a number of stacks, which are added in combination with proton exchange membrane (PEM), in
10   order to obtain separated cells for the electrochemical reactions in either the fuel cell or electrolyser. Such stacks are generally known in the art. They usually have at least 3 layers, engaging each other closely.

15   A first layer is a water- and gas-proof layer, hereafter referred to as "collector layer", and also referred to in the art as "bipolar plate". This collector layer avoids gas or water leakage from one cell to an other, and guides electrons (e⁻) to or from the cell. Therefor it is generally known to use a conductive plate, usually a graphite plate.

20

A second layer, engaging closely one side of the collector layer, is used to distribute the gasses, used or provided by the electrochemical reaction in the fuel cell or electrolyser at the proton exchange membrane (PEM), over the whole surface of the fuel cell or electrolyser.
25   This layer is hereafter referred to as "distribution layer".

A third layer, engaging closely the other side of the collector layer, is a layer, used to provide the contact between diffusion layer and PEM. At this so-called contact layer or "electrode layer", the electrochemical
30   reaction takes place, due to the presence of catalytic elements, either on the contact layer or the PEM itself. Gasses, being provided via the diffusion layer to this contact layer, are to be retained sufficiently to enable the electrochemical reaction to take place.

The contact layer, and possibly also the diffusion layer, may be made hydrophobic, e.g. by impregnation or presence of hydrophobic elements such as Teflon®, or hydrophilic.

5   Depending on the place of the stack in the fuel cell or the electrolyser, a electrochemical reaction takes place in which e⁻, protons (H^+) and a gas are consumed or provided near a PEM.

The H^+ are provided or evacuated via the PEM to the electrochemical
10   reaction. Therefor, the contact between contact layer and PEM is to be as perfect as possible, since the electrochemical reaction takes place at the catalytic layer, which is close to the surface of the PEM.

The e- are provided or evacuated via the stack of collector layer,
15   diffusion layer and contact layer. Therefor, all layers engaged are to be electro-conductive, and the resistance over the stack, and especially the contact resistance at the contacts of the several layers is to be as low as possible.

20   The diffusion layer is to spread the gas flow as much as possible over the whole surface of the contact layer, in order to use the present catalytic elements as complete as possible, providing e⁻ and H^+ over the whole surface of the PEM.

25   An example of a stack is described in WO0069003 and EP0141241.

Summary of the invention.

The present invention has as an object to provide an improved stack for
30   use in fuel cells or electrolysers, which comprises metal fibers and which has a reduced electrical resistance over its thickness, an improved property of diffusing the reaction gasses over the surface of

the PEM, against which it is to be used, and an improved contact between stack and PEM.

A stack as subject of the invention comprises at least three layers:

5
- At least one gas- and liquid impermeable metal structure (collector layer), such as a metal foil or metal plate;
- At least one first metal fiber layer (diffusion layer), comprising metal fibers;
- At least one second metal fibers layer (contact layer), comprising

10
metal fibers.

According to the present invention, the impermeable structure is sintered to one side of the diffusion layer, and the second metal fiber layer is sintered to the other side of the diffusion layer. The term layer is

15
to be understood as an essentially flat object having a thickness, which is essentially equal over the surface of the object. The properties of both metal fiber layers are chosen in such a way that the planar air permeability of the stack as subject of the invention is more than 0,5 l/min*cm, whereas the porosity of the contact layer is less than 80% or

20
even less than 75%.

A stack as subject of the invention may comprise
- a gas- and liquid impermeable metal structure (collector layer), such as a metal foil or metal plate;

25
- two first metal fiber layers (diffusion layers), comprising metal fibers;
- two second metal fiber layers (contact layers), comprising metal fibers

It is understood that at both sides of the impermeable metal structure, a

30
first metal fiber layer is sintered, and that a second metal fibers layer is sintered to each of the sides of the first metal fibers layers, which are not connected to the impermeable metal structure. The planar air permeability of the stack as subject of the invention at both sides of the

impermeable metal structure is more than 0.1 l/min*cm, whereas the porosity of the contact layers are less than 80% or even less than 75%.

With planar air permeability is meant the amount of gas which is passed through the metal fibers layers of the stack in a direction parallel to the plane of the layers. This planar air permeability is measured by taking a rectangular part of the stack, having a hight of 2.5cm. this side is hereafter referred to as short side. The other side of the rectangular sample is referred to as long side. This rectangle is clamped between two seals of equal dimension, in such a way that the sides of the sample and the sides of the seals coincide. Air is sucked using an underpressure of 200Pa at the long side of the rectangular sample over a length of 5 cm. The non-used length of the long side via which is sucked is sealed. The volume of air sucked is measured, and the permeability is expressed in l/min*cm, for which cm is the unity of length of the sample. Preferably, a length of 5 cm of the long side is used.
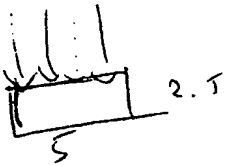
The "porosity" of a layer is expressed as

$$Porosity = 100 - Density.$$

With "density" is meant the ratio of the weight per volume of the layer over the weight of the same volume, provided 100% out of the metal alloy out of which the metal fibers are provided. The density is expressed in percentage.

Preferably the impermeable metal structure is a metal foil or metal plate, most preferably provided out of stainless steel, Ni or Ti. In case of stainless steel, preferably Fe-Ni-Cr alloys are used such as alloys of the serie AISI-300 and AISI-400, preferably AISI 316L or Ni-Cr alloys such as Inconel®.

The metal fibers used to provide the metal fibers layers are preferably stainless steel fibers, Ni-fibers or Ti-fibers. In case of stainless steel fibers, preferably Fe-Ni-Cr alloys are used such as alloys of the serie AISI-300 and AISI-400, preferably AISI 316L or Ni-Fe alloys such as Inconel®. The metal fibers may be obtained by using presently known techniques, such as bundle drawing, coil shaving or any other production technique.

Preferably all layers of the stack are provided out of the same metal or metal alloy.

The contact layer and the diffusion layer may be sintered separately, before a stack, comprising a collector layer, one or more diffusion layers and one or more contact layers, is assembled and the layers are sintered to each other. Alternatively, the diffusion layers and the contact layers, both comprising metal fibers, and the collector layer may be sintered to each other all at once during one sintering operation, either batch or continuous sintering.

The equivalent diameter of the metal fibers used to provide the first metal fiber layer or layers, so-called diffusion layer, is larger than 20µm, most preferably larger than 50µm. Possibly, more than one sheet of metal fibers is used to provide the first metal fibers layer or layers.

With "equivalent diameter of a metal fiber" is meant the diameter of an imaginary circle which has the same surface as a cross-section of the metal fiber.

Preferably, the porosity of the diffusion layer is more than 10% larger than the porosity of the contact layer. Most preferably, the porosity of the diffusion layer is more than 80%, or even more than 82%, such as more than 85% or even more than 90%. In order to obtain a preferred

stack as subject of the invention, the thickness of the diffusion layer is more than 0.5mm, most preferably more than 1mm.

Such diffusion layers provide to the largest extend the planar air permeability to the stack, and hence a superior distribution of the reaction gas over the whole surface of the adjacent contact layer or layers.

Alternatively, but also according to the present invention, a metal mesh or stretch metal foil or plate may be inserted between two sheets of metal fibers, comprised in this first metal fibers layer.

The equivalent diameter of the metal fibers used to provide the second metal fibers layer, so-called contact layer, is smaller than $30\mu m$, most preferably smaller than $10\mu m$. Possibly, more than one sheet of metal fibers is used to provide the second metal fibers layer.

The thickness of the contact layer is preferably less than 0.2mm.

Such contact layers, obtaining reaction gas via the diffusion layer over its whole surface, retains the gas sufficiently to propagate the electrochemical reaction at its reactive side, being the side of the contact layer, which contacts the adjacent PEM. Since the perpendicular air permeability of the second metal fiber layer is relatively low, a too quick consumption of reaction gas at the gas entrance of the stack is prevented, which results also in a presence of reactive gas over the whole surface of the connection layer. Possibly, the side of the contact layer, which contacts the PEM, is provided with an appropriate catalyst. Alternatively the catalyst is present on the surface of the PEM. Due to the small equivalent diameter of the metal fibers used to provide the contact layer, and the density of this layer, a very high degree of contact between contact layer and PEM may be obtained, whereas the side of the layer contacting the PEM, is relatively soft. Metal fibers projecting out of the essentially flat surface of the contact layer, will not penetrate through the PEM when used, but will bend to the contact layer surface during assembly and use of the fuel cell or electrolyser.

-7-
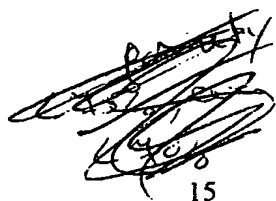
Preferably, the perpendicular air permeability of the contact layer is less
than 200 l/min*dm², most preferably even less than 150 l/min*dm².
With perpendicular air permeability is meant the amount of gas which is

5   passed through the metal fibers layer in a direction perpendicular to the
plane of the layer, measured using an underpressure of 200Pa, and
measured using methods known in the art, such as a Textester
FX3300. The planar air permeability is expressed in l/min*dm².

10   Further, the obtained stack as subject of the invention preferably has a
transversal electrical resistance of less than $30 *10^{-3}$ Ohm or even less
than $15 *10^{-3}$ Ohm, in case a stack of an impermeable metal structure,
a diffusion layer and a contact layer is provided. With transversal
resistance is meant the electrical resistance measured between a point

15   on the surface of the impermeable metal structure and the point of the
side of the connecting layer to be used against the PEM, which point is
closest to the point of the impermeable metal structure. This low
resistance is obtained thanks to the many contact points between
several fibers being sintered to each other or to the collector layer.

20   Alternatively, when at each side of the impermeable metal structure a
diffusion layer and a connection layer is provided, the transversal
electric resistance is preferably less than $30 *10^{-3}$ Ohm.

After sintering, possibly, but not necessarily, the metal fiber layers may

25   be impregnated with a hydrophobic or hydrophilic agent, e.g.
polytertrafluoethylene such as Teflon® as hydrophobic agent.

Such stacks may be used in fuel cell, where at least two stacks as
subject of the invention are used. Between a contact layer of the first

30   stack and a contact layer of a second stack, a PEM is provided. At both
sides of the PEM, necessary catalysts are present to propagate the
electrochemical reaction wanted. To the diffusion layer of the first stack,
H_2 is provided, which flows through the whole diffusion layer (due to the

high

relatively low planar air permeability of the stack). At the PEM, a
reaction as underneath is propagated:

$$H_2 \rightarrow 2\,H^+ + 2\,e^-$$
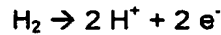
5

This side of the fuel cell is referred to as anode.
H+ is conducted through the PEM to the opposite side of the PEM,
whereas the e⁻ is drained away through the electrically conductive
connection and diffusion layer to the impermeable metal structure.

10

The e- is guided via an electrical circuit to the other impermeable metal
structure of the second stack. Again via the diffusion and connection
layer of this second stack, the e⁻ are provided to the electrochemical
reaction at this side of the PEM, being the cathode side.

15

O_2 is provided to the diffusion layer of this second stack, which is
conducted through the connection layer to the surface of the PEM.
Here a reaction takes place, using O_2, e⁻ and H⁺ (provided through the
PEM):

20

$$O_2 + 4\,H^+ + 4\,e^- \rightarrow 2H_2O$$
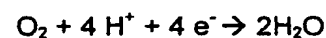
contact layers

Since the optimal planar air permeability of the stack, and the relatively
low perpendicular air permeability of the ~~connection~~ layers, the gasses

25
are spread in an optimal way over the whole PEM surface. Further,
since the low electrical transversal resistance of the stacks, the e⁻ are
conducted to the electrical circuit without a significant loss of power.
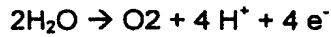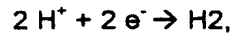
A similar benefit is made when the stacks are used in electrolysers. An
30
identical combination of at least two stacks is provided. A determined
electrical
tension is provided between the two impermeable metal structures. At
the stack with a positive tension on its impermeable metal structure,
H_2O is provided, which reacts at the PEM surface as:

$$2H_2O \rightarrow O2 + 4\,H^+ + 4\,e^-$$

the H^+ is conducted through the PEM to the other side of the PEM, whereas the e^- is conducted via the metal fiber layers to the impermeable metal structure. O_2 is easily evacuated since the high planar air permeability of the diffusion layer.

At the other side, a reaction takes place:

$$2\,H^+ + 2\,e^- \rightarrow H2,$$

Where H^+ is provided via the PEM and the e^- are provided via the impermeable metal structure (being negative pole) and the metal fiber layers. H_2 is easily evacuated due to the high planar air permeability of the diffusion layer.

Brief description of the drawings.

The invention will now be described into more detail with reference to the accompanying drawings wherein

- FIGURE 1 shows schematically a stack as subject of the invention;
- FIGURE 2 shows schematically another stack as subject of the invention;
- FIGURE 3a, FIGURE 3b and FIGURE 3c show schematically a test set up to measure the planar air permeability of a stack;
- FIGURE 4 shows schematically a fuel cell as subject of the invention;
- FIGURE 5 shows schematically an electrolyser as subject of the invention.

Description of the preferred embodiments of the invention.

An embodiment of a porous stack as subject of the invention is shown in Figure 1.

The stack 10 comprises a gas- and liquid impermeable metal structure, so-called collector layer 11. In the embodiment shown in Figure 1, a metal plate provided out of stainless steel with alloy AISI 316L is used. The collector layer has a thickness of 0,4 mm.

5

To one side of this collector layer, a diffusion layer 12, being a sintered metal fiber layer consisting out of stainless steel fibers (Alloy AISI 316L) with equivalent diameter of 22 µm is provided. This diffusion layer 12 has a thickness of 1,7mm and has a porosity of 85%.

10

(90%

The fibers are obtained by coil shaving process, which provides to the fibers a substantially rectangular cross-section, have a large side of approximately 25 µm and a short side of approximately 15 µm

15

To the side of the diffusion layer, which does not contact the collector layer, a second metal fiber layer 13, being the contact layer, is provided.

20 This contact layer 13 consists of sintered bundle drawn stainless steel fibers (Alloy AISI 136L) having an essentially circular cross-section with an equivalent diameter of 8 µm. This contact layer 13 has a thickness of approximately 0,2 mm and a porosity of preferably 70%. Alternatively a porosity of 85% may be used. -

25

An alternative stack 20 as subject of the invention is shown in Figure 2. This embodiment comprises a collector layer 21, being identical to the collector layer of Figure 1.

30 At both sides of the collector layer 21, a diffusion layer 22a and 22b is provided, each diffusion layer 22a and 22b being identical as the diffusion layer of Figure 1.

Similar as in Figure 1, a contact layer 23a and 23b is provided to the sides of diffusion layer 22a and 22b. These contact layers 23a and 23b are identical as the contact layer of Figure 1.

5 The planar air permeability of stack 10, and of both sides 24a and 24b of stack 20, are measured as shown in FIGURE 3a, FIGURE 3b and FIGURE 3c.

As shown in FIGURE 3a, a rectangular sample (301) of a stack, having a long side 302 and a short side 303 is clamped between two seals 304

10 by means of two clamps 305. The sealant material of seal 304 is preferably a PU foam, having a thickness of approximately 10mm when not pressed. The clamps 305 are preferably made out of metal. All parts are kept together by closing means 306, e.g. a clip.

This assembly is placed on a sealant 307, which is provided with a hole

15 (see figure 3b and 3c), located underneath the long side 302. The dimension 308 and of the assembly is at least 2 cm larger than the diameter 310 of the hole.

The height of the long side 302 is taken 2.5cm.

In a section according to AA' in FIGURE 3b, and in the section

20 according to BB' in FIGURE 3c, it is shown that the assembly is places over the opening of a sucking device 312, e.g. a Textest FX3300, having a suction opening with a diameter 311, which is at least equal to the diameter of the opening in the sealant 307, but which is smaller than the dimension 308.

25 Air is sucked in direction 313 using an underpressure of 200Pa through the sample 301 of the stack, as indicated with arrow 314. The volume of air sucked is measured per minute, and the air permeability is expressed as the volume per minute and per length unit of the diameter of opening 310.

30

The planar air permeability of stack 10 and of both sides 24a and 24b of stack 20, as described above and shown in FIGURE 1 and 2, is

liter per minute for a length 310 of 5cm. The planar air permeability thus is 0.16l/min*cm.

0.8

It was found that the porosity of contact layer 13, 23a or 23b does not

5   make significant changes to this planar air permeability.

The perpendicular air permeability of the contact layer 13, 23a and 23b is measured to be 123 l/min*dm² for a contact layer of 70% porosity.

10   An electrical resistance may be measured over the stack 10 or over both sides of stack 20, being 9.2 * 10^{-3} Ohm.

The stacks 10 or 20 as subject of the invention may be used in an electrolyser or in a fuel cell, as shown in Figure 4 and Figure 5.

15 .

Figure 4 shows a fuel cell 40, comprising several stacks 10 and 20, separated from each other using proton exchange membranes 41, between the contact layers of the stacks 10 or 20 and the PEM 41, appropriate catalysts are provided.

20

O_2 or H_2 is provided to the stack in such a way that at both sides of the PEM, an electrochemical reaction takes place. The e⁻ are collected through the contact and diffusion layers by the collector layers.

25   The collector layers are connected to each other via an appropriate electrical connection device 42, which provided electrical current to be used by an electrical device or to a battery 43.

In Figure 5, two stacks 10 are separated from each other by means of a
30   catalytically coated PEM 51.

An electrical tension is provided by a tension source to the collector
layers of the electrolyser. H_2O being provided to the electrolyser, reacts
electrochemically, providing O_2 and H_2.

CLAIMS

1. A stack comprising an impermeable metal structure, at least one first metal fiber layer and at least one second metal fiber layer, said impermeable metal structure being sintered to one side of said first metal fibers layer, said second metal fibers layer being sintered to the other side of said first metal fibers layer, characterized in that the planar air permeability of said stack being more than 0.1 *0,5* l/min*cm, the porosity of said second metal fiber layer being less than 80%, *preferably (75 %)*

2. A stack as in claim 1, said stack comprising two first metal fiber layers and two second metal fiber layers, said first metal fiber layers being sintered each to one side of the impermeable metal structure, said second metal fiber layers being sintered to the other sides of said first metal fiber layers.

3. A stack as in claim 1 or 2, said first metal fiber layers having a porosity of more than 80% , *preferably > 85 %*

4. A stack as in claim 1 to 3, said second metal fiber layers having a perpendicular air permeability of less than 150 l/min*dm².

5. A stack as in claim 1 to 4, said first metal fiber layers comprising fibers with equivalent diameter of more than 20μm, *preferably > 50μ*

6. A stack as in claim 1 to 5, said second metal fibers layers comprising fibers with equivalent diameter of less than 30μm, *preferably 10μm*

7. A stack as in claim 1 to 6, said first metal fiber layers having a thickness of more than 0.5mm

8. A stack as in claim 1 to 7, said second metal fiber layers having a thickness of less than 0.2mm

9. A stack as in claim 1 to 8, said stack having a transversal electric resistance less than $30*10^{-3}$ Ohm

10. A stack as in claim 1 to 9, said impermeable metal structure being a metal plate.

11. A stack as in claim 1 to 9, said impermeable metal structure being a metal foil.

12. A stack as in claim 1 to 11, said metal fibers being stainless steel fibers.

13. A stack as in claim 1 to 11, said metal fibers being Ni-fibers or Ni-alloy

14. A stack as in claim 1 to 11, said metal fibers being Ti-fibers

15. A stack as in claim 1 to 14, said metal fibers having the same alloy of said impermeable metal structure.

16. A fuel cell, comprising stacks as in claim 1 to 15.

17. An electrolyser, comprising stacks as in claim 1 to 15.

18. The use of a stack as in claim 1 to 15 in fuel cells or electrolysers.

ABSTRACT

A stack to be used in a fuel cell or an electrolyser, comprises an impermeable metal structure, at least one first metal fiber layer and at least one second metal fiber layer. The impermeable metal structure is sintered to one side of said first metal fibers layer and the second metal fibers layer is sintered to the other side of said first metal fibers layer. A planar air permeability of said stack of more than 0.1 l/min*cm is provided, whereas the porosity of said second metal fiber layer being less than 80%.

Dossier: 5583

Titel: porous metal stack for fuel cells or electrolysers.

5   Aanvrager: NV BEKAERT

Uitvinders: Anaf

                    x

                    x

10

                    Losfeld

                    x

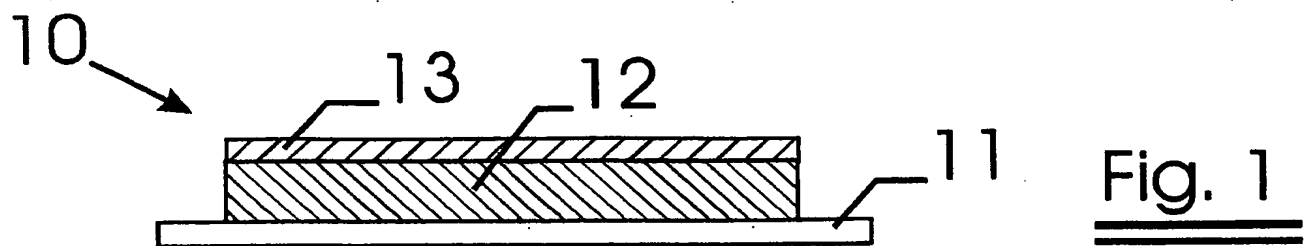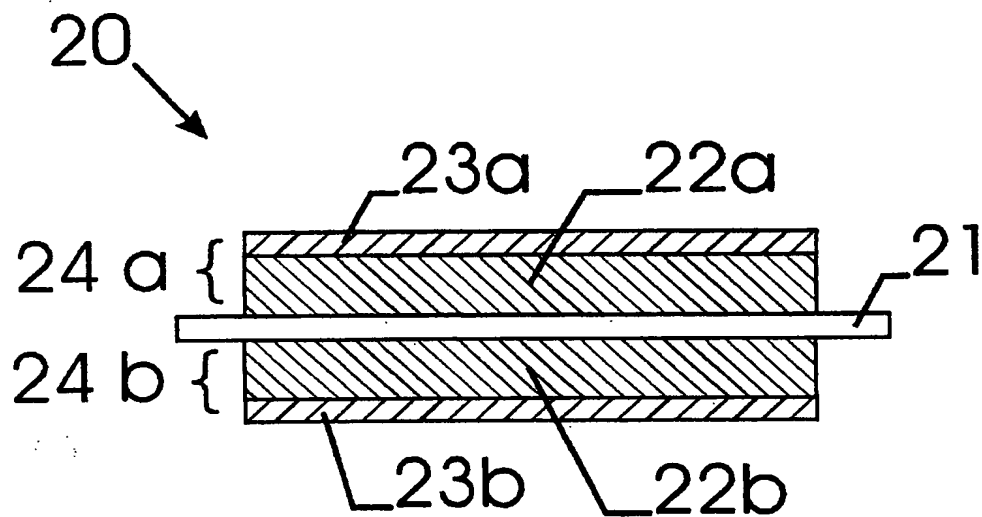   x

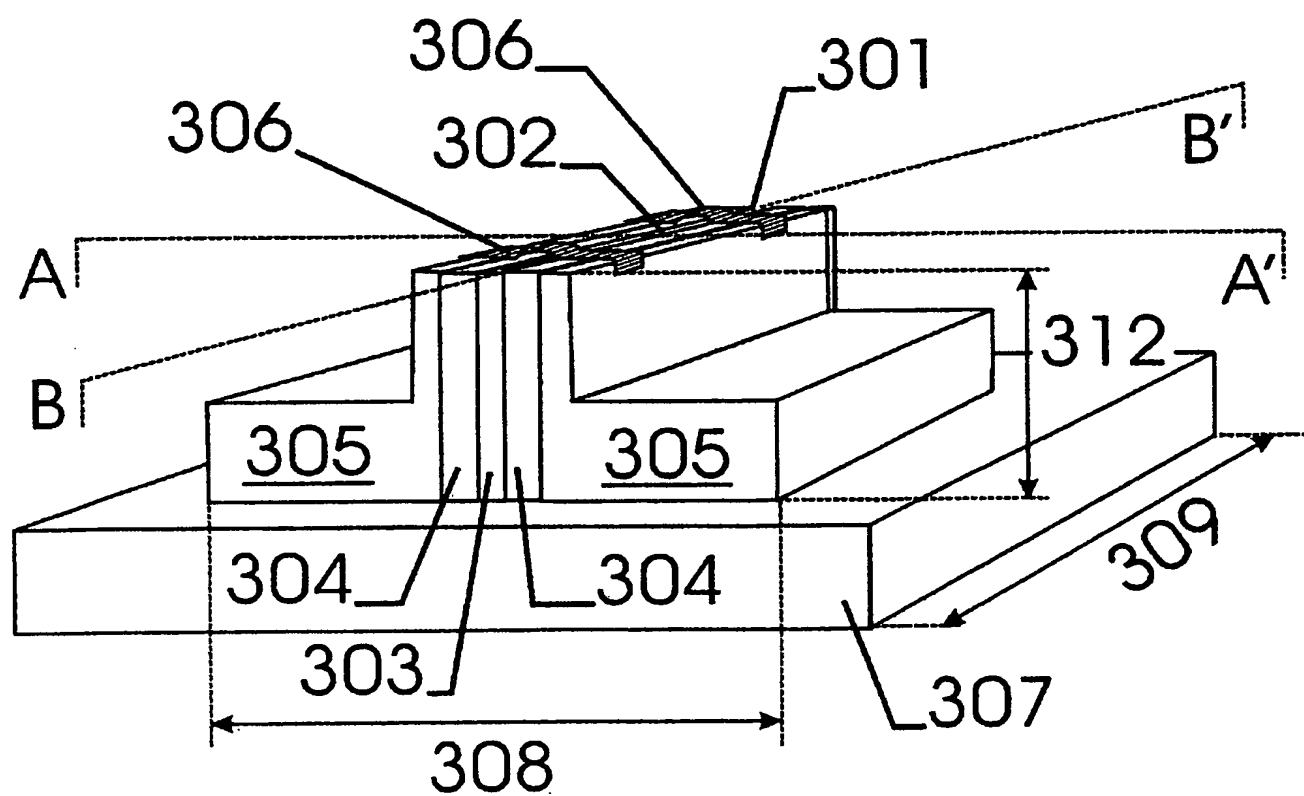15   x

                    x

                    x

Figuur abstract: figuur 1

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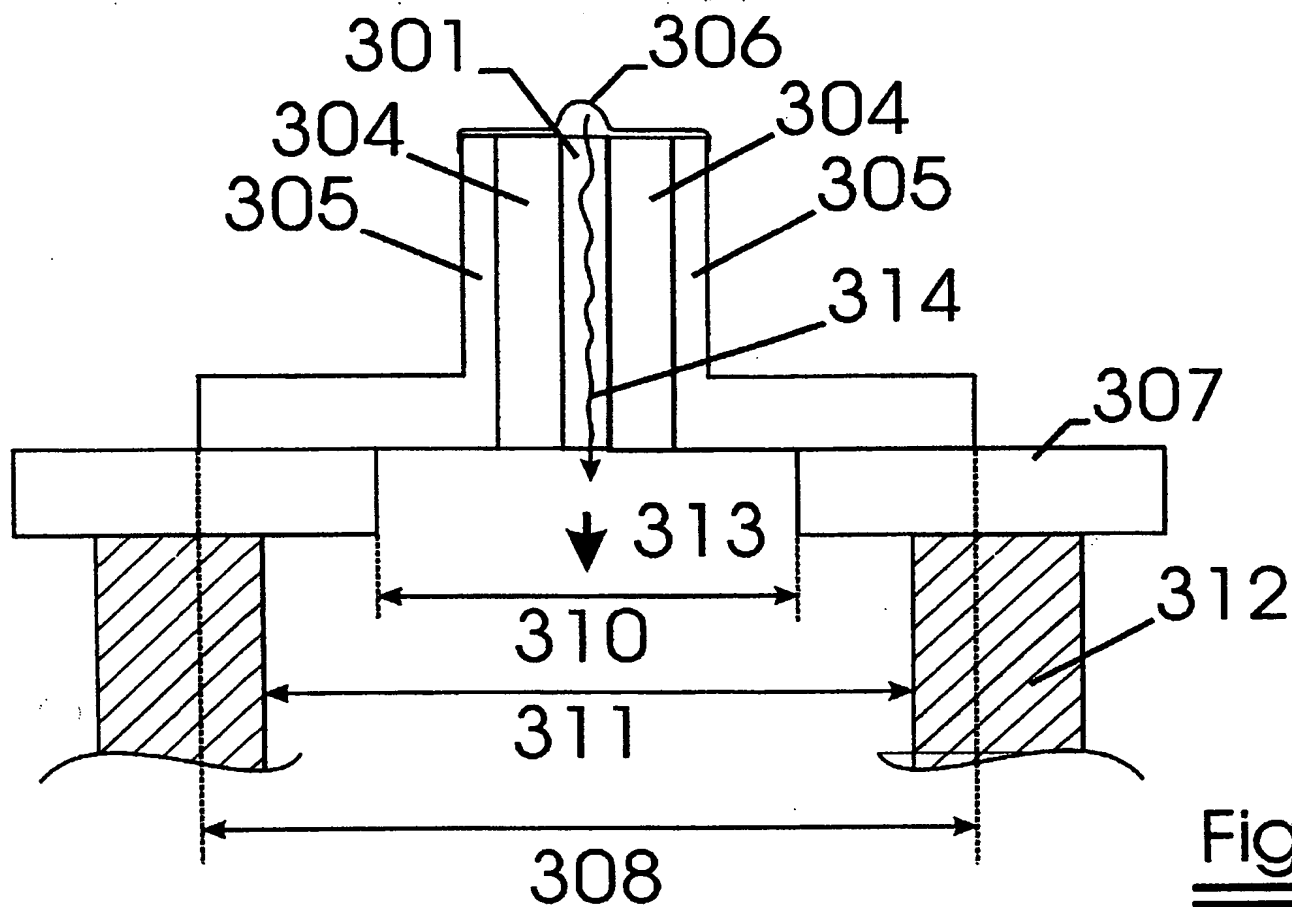11   Fig. 1

20

23a 22a

24 a {

21

24 b {

23b 22b Fig. 2

306 301

306

302

B'

A   A'

B

312

305 305

304 304

303

309

308   307

Fig. 3a

Fig. 3b

Fig. 3c

Fig. 4

Fig. 5